# Beta decay properties of  $67,68$ Se and the astrophysical rp-process path

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The <sup>67</sup>Se and <sup>68</sup>Se isotopes near the proton drip line were produced by spallation of zirconium with 600 MeV protons at CERN. Beta decay properties were determined for the first time using the selectivity of molecular ion beams at the on-line mass separator ISOLDE. Chemically selective production was obtained through separation of the molecular ion  $COSe^+$ . The half-lives of  ${}^{67}Se$  and <sup>68</sup>Se were measured as  $107 \pm 35$  ms and  $35.5 \pm 0.7$  s respectively. From  $\beta-\gamma$  and  $\gamma-\gamma$  coincidences, and conversion-electron measurements, a decay scheme was obtained for  ${}^{68}$ Se. For the  ${}^{68}$ As ground state and several excited states, spin and parity are unambiguously determined from  $\beta$ -decay rates and electromagnetic multipolarity assignments. In addition to the strong Fermi branch between the mirror ground states of <sup>67</sup>Se and <sup>67</sup>As ( $J^{\pi} = (5/2)^{-}$ ,  $T_z = \pm 1/2$ ), a GT branch to an excited state in <sup>67</sup>As is observed. Implications of <sup>67</sup>Se and <sup>68</sup>Se properties in the  $rp$ -process path are considered.

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### I. INTRODUCTION

The heaviest even-even self-conjugated  $N = Z$  nuclei for which the  $\beta$  decay has been observed are <sup>72</sup>Kr [1] and  ${}^{76}Sr$  [2,3]. Due to the difficulty of producing the Se isotopes selectively, the  $68$ Se decay properties have so far been unknown despite many experimental attempts [4—7] although detailed results for the  $69$ Se decay [8] could be obtained.

In-beam studies performed up to  $84$ Mo ( $N = Z = 42$ ) [9] have revealed large prolate deformations for  ${}^{76}Sr$  and 80Zr, in agreement with calculations of Möller and Nix [10]. Detailed in-beam  $\gamma$ -ray measurements on  $^{69}$ Se [ll—13] have given evidence for an oblate deformation. For <sup>68</sup>Se [14], a 2<sup>+</sup> candidate at  $E_x = 854$  keV does not appear consistent with the predicted large oblate deformation [15].  ${}^{68}Se$  and  ${}^{67}Se$  were identified as  ${}^{78}Kr$ beam fragments [16] and recently the  $\beta$ -delayed proton emission of <sup>65</sup>Se was observed using a fusion-evaporation reaction [17].

Astrophysical interest can be pointed out for <sup>68</sup>Se and  $67$ Se. These nuclei are involved in the  $rp$  process proposed by Wallace and Woosley [18]. This process, being responsible for the production of n-deficient isotopes of elements beyond Ni is suggested to occur in x-ray bursts as the outcome of hydrogen and helium explosions on the surface of neutron stars. In particular, binding energies, half-lives, and  $\beta$ -strengths of  $67,68$ Se should be known, in order to assess the importance of these nuclides in the rp process [18,19].

In this work, we present results obtained for the  ${}^{68}Se$ and  $67$ Se  $\beta$  decay at the ISOLDE on-line separator at CERN. Preliminary data has been reported in [20].

#### II. EXPERIMENTAL PROCEDURE

With the intent to observe the  $\beta$  decay of exotic selenium isotopes, we performed an experiment at the ISOLDE on-line separator using the 600 MeV proton beam of the SC, bombarding a  $ZrO<sub>2</sub>$  target associated with a plasma ion source. It was proved [21] from previous tests that good yields could be obtained for selenium from a  $ZrO<sub>2</sub>$  felt target. It was also shown that controlled addition of  $O_2$  gas led to the production of  $\mathrm{COSe}^+$  molecular ions, thus facilitating chemically selective production ofradioactive Se isotopes in a sideband shifted by 28 mass units. The experimental setup for observing the decay of the mass-separated isotopes was arranged around the collection point located on the Mylar ribbon of a tape transport system. It consists of a  $4\pi$ -beta counter surrounding the collection point and detectors for  $\beta-\gamma$  and  $\gamma$ - $\gamma$  measurements. A small Ge(Li) counter (relative efficiency  $2\%$ ) was used to detect the low-energy gamma rays and two Ge counters (relative efficiencies 33 and  $70\%$ ) to perform  $\gamma$ - $\gamma$  coincidence measurements. The lifetime, or an upper limit, associated with each low-energy transition was determined using the delayed  $\gamma$ - $\gamma$  coincidence technique. Conversion-electron measurements were also carried out with a cooled Si detector (1 mm thick) in order to obtain the multipolarity of low-energy transitions.

### III. EXPERIMENTAL RESULTS

## A. The  ${}^{68}Se \beta$  decay

Due to the presence of isobars, on-line mass separation of the elementary ions is not sufficient to identify the most exotic Se isotopes. Figure 1(a) shows a  $\beta$ -coincident  $\gamma$  spectrum taken at mass  $A = 68$ . We observe the  $\beta$ decay of  ${}^{68}Cu$ ,  ${}^{68}Ga$ , and  ${}^{68}As$ . We could not assign lines to the <sup>68</sup>Se decay, screened by the high production rate of these isobars.

When the target was operated with an  $O<sub>2</sub>$  leak, selective production of Se isotopes as the molecular ion COSe+ was obtained. Gamma-ray spectra taken at mass separated  $A = 97$  (CO<sup>69</sup>Se<sup>+</sup>) reveal a pure production of  $69$ Se, whose  $\beta$  decay is well known [8]. The yield achieved for the <sup>68</sup>Se production as  $CO^{68}Se^+$  ( $A = 96$ ) at our experimental station was about 300 atoms/s using a proton



FIG. 1. (a) Portion of the  $\beta$ -coincident  $\gamma$  spectrum taken at  $A = 68$ . (b) Portion of the  $\beta$ -coincident  $\gamma$  spectrum taken at  $A = 96 \, (\text{CO}^{68}\text{Se}^+)$ . The peaks attributed to the <sup>68</sup>Se decay are labeled with the corresponding  $\gamma$ -ray energy in keV.



FIG. 2. Coincidence spectra on the 111 and 161 keV lines (background has been subtracted).

intensity of 2.5  $\mu$ A. A portion of the  $\beta$ -coincident  $\gamma$  spectrum taken at  $A = 96$  is shown in Fig. 1(b). Most of the observed  $\gamma$  lines were assigned to the  $\frac{68}{10}$ Se decay. The line at 98 keV which belongs to the <sup>69</sup>Se decay is seen, due to limited mass resolution. The line at 64 keV comes from the <sup>80</sup>Rb decay present as a compound RbO<sup>+</sup> ( $A = 96$ ). The remarkable selectivity obtained at  $A = 96$  for the  $68$ Se production is illustrated by comparing Figs. 1(a) and 1(b).

 $\gamma$  spectra were recorded in a multispectrum mode (time-shifted  $4 \times 20$  s). The decay of the 111, 161, 192, 265, and 314 keV  $\gamma$  lines was analyzed. The weighted mean value of the results leads to the half-life of  $T_{1/2}$  =  $35.5 \pm 0.7$  s for <sup>68</sup>Se. The <sup>68</sup>Se decay scheme established from  $\gamma$ - $\gamma$  coincidences (Fig. 2),  $\gamma$  intensities and energy sums, is shown in Fig. 3. The sequence of the 314—111



FIG. 3. Experimental  $^{68}$ Se  $\beta$ -decay scheme. Beta intensities and logft values are indicated. The  $Q_{EC}$  value is from [22].

TABLE I. Energy, intensity, and assignment of  $\gamma$  transitions in the <sup>68</sup>Se  $\beta$  decay.

$E_{\gamma}$	$I_{\gamma}$	Transition
(keV)	$(\rm relative)$	(keV)
$49.5 \pm 0.3$	$63 + 10$	$161 - 111$
$72.6 \pm 0.5$	$188 + 27$	$426 - 353$
$111.4 \pm 0.2$	$1430 \pm 100^a$	$111 - 0$
$160.8 + 0.2$	$808 + 43$	$161 - 0$
$192.2 \pm 0.5$	$198 + 23$	$353 - 161$
$202.7 + 0.5$	$59 + 18$	$364 - 161$
$265.0 \pm 0.3$	$467 + 39$	$426 - 161$
$314.5 \pm 0.3$	$1133 + 69$	$426 - 111$
$352.6 \pm 0.5$	$130 + 13$	$353 - 0$
$426.1 \pm 0.4$	$134 + 29$	$426 - 0$

 ${}^{\rm a}I_{\gamma} + I_{\rm e}$ , taking into account the measured total conversion coefficient  $\alpha = 0.43 \pm 0.10$ .

keV cascade was deduced from the delayed coincidence results. The intensities of the  $\beta$  branches are deduced from the imbalance of the  $\gamma$  intensities connected with each level.  $Q_{EC} = 4730 \pm 310$  keV [22] has been used for the  $\beta$  transition rate determination. Ground-state spin  $J = 3$  for <sup>68</sup>As is taken from the literature with no indication on parity [23].

The energy and intensity values for the  $\gamma$  transitions are given in Table I along with the corresponding assignments in  ${}^{68}\text{As}$ . Table II shows the  $\gamma$ -ray branching ratios in <sup>68</sup>As. Excitation energies,  $\beta$  intensities, and the corresponding  $\log ft$  values are listed in Table III. We can assign  $J^{\pi} = 1^{+}$  to four excited states on the basis of logft values.

Associated with the 111 keV  $\gamma$  ray we observed a Kelectron line (Fig. 4). The comparison of the two intensities yields a value of the total conversion coefficient  $\alpha = 0.43 \pm 0.10$ , compatible only with an E2 or M2 transition. From the delayed coincidence measurement we deduced the half-life of  $T_{1/2} = 107^{+23}_{-16}$  ns for the 111<br>keV level. Taking into account our measured values ( $T_{1/2}$ ) and  $\alpha$ ) we obtain a strength of either  $B(E2) = 13 \pm 3$ W.u. or  $B(M2) = 720 \pm 160$  W.u. for the 111 keV transition. The recommended upper limits [24] allow only the  $E2$  transition. As a consequence, the parity of the  $^{68}$ As ground state is determined as positive.

TABLE II.  $\gamma$ -ray branching ratios in <sup>68</sup>As.

$E_i$	$E_{f}$	Gamma branching
(keV)	$(\mathrm{keV})$	ratio $(\%)$
111	0	100
161	0	$93 + 2$
	111	$7 + 2$
353	0	$40\pm4$
	161	$60 + 4$
364	161	100
426	0	$7 + 2$
	111	$59 + 3$
	161	$24\pm2$
	353	$10\pm2$



FIG. 4. Conversion-electron spectrum for low-energy transitions in <sup>68</sup>As. The low-energy peaks at 10.5 and 14.1 keV are x rays from As and Sr, respectively (the line at 24 keV is a contamination of the detector).

As the direct population of the 161 keV level is uncertain,  $\beta$  decay cannot give information on its spin; the experimental limit on its half-life,  $T_{1/2}$  < 10 ns, is not short enough to give additional constraint. However, the relative intensities of the  $\gamma$  transitions of energies 49.5 and 160.8 keV can be obtained only with the spin value  $J = 2$ , the two transitions being dipole of the same type (both  $M1$  or both  $E1$ ).

None of the excited states found in  ${}^{68}$ As by  $\beta$  decay can be related to the <sup>68</sup>As levels reported previously in heavy-ion reaction studies [23]. This explains the difficulty encountered in the identification of <sup>68</sup>Se in former studies.



FIG. 5.  ${}^{67}Se$   $\beta$ -counting rate during the collection and the decay phases. Experimental values are stored in time bins of 30 ms. The solid line corresponds to a calculated fit with  $T_{1/2} = 88$  ms.



FIG. 6. Experimental  $^{67}$ Se  $\beta$ -decay scheme. Beta intensities, logft values, and experimental values of the GT matrix elements are reported. The  $Q_{EC}$  value is from [22].

## B. The  $67$ Se  $\beta$  decay

The separator was set on mass  $A = 95$  corresponding to the molecular ion  $CO^{67}Se^+$ . The production rate measured by  $\beta$  counting was about 16 atoms/s. We supposed that the whole measured beta activity was due to <sup>67</sup>Se, as was the case for  $A = 97$  and 96, where the major intensity came from <sup>69</sup>Se and <sup>68</sup>Se because of the selectivity of the molecular ion.

We determined the half-life of <sup>67</sup>Se by collecting the radioactive beam during 120 ms every 1.2 s. The  $\beta$  activity was measured in a  $4\pi\beta$  plastic counter, in a multiscale mode during collection and decay with time intervals of  $\Delta t = 30$  and 100 ms. Figure 5 shows the  $\beta$  counting rate during the collection and the decay with  $\Delta t = 30$ ms; a global fit to this curve yields  $T_{1/2} = 88$  ms for the half-life. A weighted mean value of the two results (for  $\Delta t = 30$  and 100 ms) leads to  $T_{1/2} = 107 \pm 35$  ms for the <sup>67</sup>Se half-life.

In the  $\beta$ -coincident  $\gamma$  spectrum we observe mainly a strong  $\gamma$  line at 511 keV corresponding to the annihilation radiation of the  $\beta^+$  transition connecting the ground states of the mirror nuclei 67Se and 67As. In



FIG. 7.  $rp$ -process path at  $T_9 = 1.5$  and  $\rho = 10^6$  g cm<sup>-3</sup> in the region of interest as proposed by Champagne and Wiescher [19].

TABLE III.  $\beta$  intensities and logft values in the <sup>68</sup>Se  $\beta$ decay to <sup>68</sup>As excited states.

$E_{\bm{x}}$	$I_{\beta}$	$\log ft$
(keV)	(per 100 decays)	
$111.4 \pm 0.2$	$10.1 + 4.7$	$5.3^{+0.3}_{-0.4}$
$160.8 \pm 0.2$	< 6.1	> 5.0
$352.8 \pm 0.4$	$6.0 + 1.4$	$5.4 \pm 0.3$
$363.5 \pm 0.5$	$2.6 \pm 0.7$	$5.8 + 0.3$
$425.9 \pm 0.2$	$81.3 \pm 4.9$	$4.2 \pm 0.2$

addition, a weak  $\gamma$  ray appears at 352 keV, not observed by Lang et al. [25] in the study of  $67$ As excited states by fusion-evaporation reactions. To calculate the  $\log ft$ values (Fig. 6), we used the evaluated <sup>67</sup>Se-<sup>67</sup>As mass excess  $(Q_{EC} = 10.15 \pm 0.22 \text{ MeV})$  from [22]. The experimental values of the GT matrix elements reported in Fig. 5 were obtained using the procedure and values for constants given in [26]. For the  $\beta$  transition to the excited state ( $E_x = 352$  keV) the Fermi contribution is negligible, while for the ground-state transition the GT value  $(0.38 \pm 0.13)$  is determined after deduction of the superallowed Fermi contribution. For this transition the experimental value of the GT matrix element is found to be about 50% lower than the value evaluated in the framework of the single-particle model. This reduction is comparable to the one  $(35\%)$  observed for <sup>71</sup>Kr [27] and also to those previously measured for the  $f_{1/2}$  shell mirror nuclei.

## C. Se isotopes involved in the  $rp$ -process reaction path

Reaction flows of the rp process are evaluated from network calculations where the nuclear properties and temperature and density conditions are taken into account. The reaction network used by Wallace and Woosley [18] has been updated [19] and the reaction flow pattern calculated for a temperature  $T_9 = 1.5$  K and a proton density  $\rho = 10^6$  g cm<sup>-3</sup>. The results are presented in Fig. 7. which is taken from [19]. Winger et al. [28] have recently demonstrated the <sup>65</sup>As stability against proton decay. This allows the reaction flow to continue towards higher masses including the selenium isotopes of mass  $A = 66$ -68. <sup>66</sup>Se is still unobserved, its decay is expected mainly to proceed through the superallowed  $0^+ \rightarrow 0^+$  transition with a correspondingly short half-life.

The half-lives we have measured for <sup>67</sup>Se  $(T_{1/2} = 107 \pm$ 35 ms) and for <sup>68</sup>Se ( $T_{1/2} = 35.5 \pm 0.7$  s) are consistent with the pattern proposed for the  $rp$  process in this mass region [19]. For <sup>67</sup>Se the half-life is short and we have mainly  $\beta$  decay to <sup>67</sup>As. For <sup>68</sup>Se the long half-life allows proton capture within the relevant time scale. The next candidate for  $rp$ -process termination point will be  ${}^{72}\text{Kr}$ unless<sup>73</sup>Rb were a (particle-bound) beta emitter of short half-life (few 100 ms).

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